CATHODE MATERIALS FOR SOLID OXIDE FUEL CELLS

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The state-of-the-art Westinghouse tubular solid oxide fuel cells (SOFCs) operate at 1000°C and use Lanthanum Strontium Manganite (LSM) as the cathode material, yttria stabilized zirconia (YSZ) as the electrolyte, and Ni-YSZ cermet as the anode. Under normal operating conditions the overpotential at the cathode has a value around 60mV due to the low oxide ion conductivity of the manganite. With this in mind, one of our main objectives is to increase the ionic conductivity of LSM through a strategy of selective B-site doping.

In our study, gallium was chosen as the B-site dopant for LSM. A series of compositions of LSM doped with various amounts of Ga were synthesized by the glycine nitrate method. The concentration of both dopants $(La_{1-x}Sr_x)(Mn_{1-y}Ga_y)O_3$ was varied ($0 < x < 45$mol %; $0 < y < 60$mol % respectively) in order to obtain single perovskite phase material. It was found that the single perovskite phase only existed in two doping ranges: x = 40 ~ 50mol % and $y < 30$mol %; and $x < 20$mol % and $y \leq 40$mol %. The unit cell volume of the single phase compositions was calculated by Rietveid refinement of the X-ray data. When the Ga concentration was less than 20mol %, little change of the unit cell volume was observed, consistent with the similar atomic radii of gallium and manganese cations. Above 20mol % Ga, the unit cell volume increased with increasing gallium concentration. This was interpreted as that at low gallium concentration, the dopant was probably randomly distributed within the lattice, while at higher concentrations, partial ordering or oxygen loss was likely to be occurring.

Single phase materials were studied as air electrode by using both impedance spectroscopy and DC polarization techniques. The sintering behavior was studied by measuring cathodic polarization resistance (R_c) under open circuit potential (OCP) condition in order to obtain optimal firing temperature for the electrode formation. At the optimal firing temperature (1100°C), the effect of Ga dopant concentration (ranging from 10 to 40mol %) on the cathode behavior was examined by determining R_c under OCP. The R_c obtained under OCP depends strongly on the concentration of Ga dopant. R_c increases with increasing Ga content, indicating that higher concentrations of Ga doping are not desirable, and implying that a lower Ga doping would yield better cathode performance.

At relatively low Ga concentrations ($0 \leq y \leq 10$mol %), the polarization curves of doped LSM were determined. The experimental results have shown that a suitable amount of Ga doping can lower the cathodic overpotential of LSM. The optimum Ga doping concentration is around 5mol %, as shown in Figure 1. In future experiments this effect will be verified in full cell tests.

In parallel with improving the properties of lanthanum manganite, ANL is exploring new cathode materials for low temperature SOFCs. Several classes of perovskite-related materials are being explored, i.e. ferrates, nickelates, and cobaltates. In addition, some layered oxide and composite structured materials are being explored. Two separate electrolytes were chosen for electrically screening these materials in two temperature regimes. For temperature below 650^{o}C, the testing was performed with a gadolinia doped ceria electrolyte (CGO), while in the range of 650 ~ 850^{o}C, YSZ was used.

Various compositions of cobaltates, ferrates, and nickelates were prepared using the glycine nitrate method. Only single phase perovskites confirmed with XRD were electrochemically characterized as cathodes in air using impedance analysis. The effect of temperature on the electrode polarization resistance was examined and the activation energy for air reduction on various cathodes was calculated. A series of XRD tests were also performed to study the reactivity of these potential cathodes with the CGO and YSZ electrolytes.

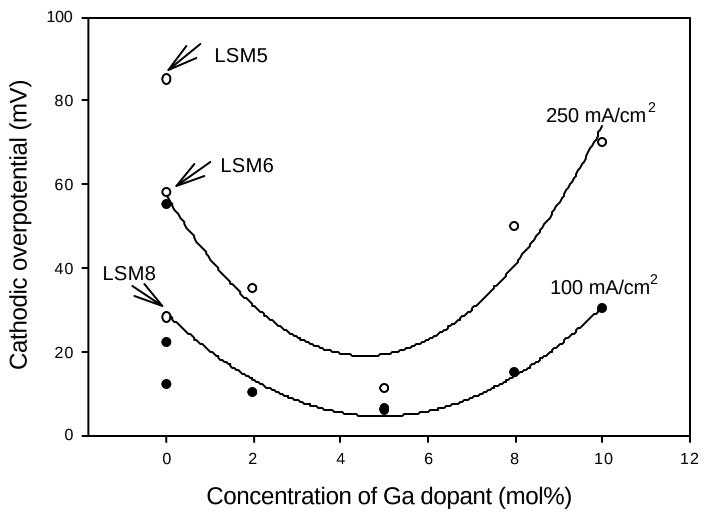

Figure 1. Effect of Ga dopant concentration on the cathodic overpotential of LSM cathodes. LSM5 = $(La_{0.55}Sr_{0.45})_{0.99}MnO_3$, LSM6 = $La_{0.6}Sr_{0.4}MnO_3$, LSM8 = $La_{0.8}Sr_{0.2}MnO_{3.}$